

large interface allows good access of reactants to the channels of the zeolite. As a result, the metal-containing macrostructures can have enhanced performance when used in many commercial applications.

The macrostructures are made by: (a) forming a mixture of porous organic ion exchanger and a synthesis gel which is capable of forming said porous inorganic material; (b) converting the synthesis gel within the pore space of said porous organic ion exchanger to the porous inorganic material; and (c) removing the porous organic ion exchanger to obtain the macrostructures. The metal can be added to the macrostructures during their formation, after the macrostructures have been formed, or both.

The macrostructures include at least one metal and comprise a three-dimensional network of self bound particles of porous inorganic material. Preferably, the porous inorganic material comprises zeolite. The particles are joined together to form a three-dimensional interconnected network of pores and occupy less than 75% of the total volume of the macrostructures. Thus, more than 25% of the total volume of the macrostructures is unoccupied by the particles of porous inorganic material. The porosity of the three-dimensional interconnected network is external to the particles of the macrostructures.

Usually the porosity of the three-dimensional interconnected network of Applicant's macrostructures is not uniform and the size of the particles is usually small, e.g., less than 500 nm. The pores of the macrostructures provide openings for conducting reactants and products between the exterior surface of the macrostructures and the inorganic oxide particles within the macrostructure.

Problem Solved by Applicants' Invention

Porous inorganic materials, e.g., zeolite, are used in a number of commercial applications, e.g., catalysts or catalyst supports for chemical processes.

Zeolites are crystalline microporous inorganic materials. One family of zeolite is aluminosilicate zeolite. Aluminosilicate zeolites are crystalline microporous inorganic materials that consist of a 3-dimensional structure of $[\text{SiO}_4]^{4-}$ and $[\text{AlO}_4]^{5-}$ coordination polyhedra linked by all of their corners. Within the zeolite structure are channels or pores which may be interconnected by a number of still smaller channels or pores. The dimensions of the channels are such as to allow adsorption of molecules with certain dimensions while rejecting those with larger dimensions.

Before using zeolite crystals in many of the commercial applications, it is usually necessary that they be shaped into macrostructures that are resistant to mechanical attrition, i.e., resistant to the formation of small particle fines, e.g., particles having a size of less than 20 microns.

For many applications, it is important that the macrostructures have a large interface between the gas or liquid phase of the reactants and products and the solid phase of the porous inorganic material of the macrostructures. The accessibility of the surface area of the macrostructure to reactant chemisorption and product desorption as well as the amount of surface area of macrostructures are significant factors in their effective use. Generally, macrostructures with a high surface area and having good accessibility to its surface area are more active than macrostructures without these characteristics. In the case of macrostructures comprised of zeolite, it is especially important that the macrostructures have a large interface so that the reactants have access to the channels/pores of the zeolite.

Many of the prior art techniques used to make macrostructures involve mixing zeolite with binding material to form an extrudable mass and then extruding the extrudable mass to form extrudates. Macrostructures prepared by this technique can have poor porosity because the binding material may block access to the pores of the zeolite and prevent hydrocarbons from entering the pores of the zeolite and undergoing conversion within the channels. A further problem in the use of amorphous binder material is that it may itself catalyze undesirable reactions which can result in the formation of undesirable products. Still further, the binder material can dilute the active phase of the catalyst and thus affect activity of the catalyst.

Claims 1-4, 6-12, 14-24, 26-28, 33-36 and 38-41 stand rejected under 35 USC 102(b) as anticipated by or, in the alternative, under 35 USC 103(a) as obvious over U. S. Patent 4,680,170 (Lowe et al.). In addition, Claims 1-41 stand rejected under 35 USC 103(a) over Lowe et al. further in view of U.S. Patent 6,160,191 (Smith et al). These rejections are specifically traversed as the invention, as defined in presently pending Claims 1 through 41, is submitted to be patentable over Lowe et al. alone, and in combination with Smith et al.

Lowe et al. is not concerned making macrostructures. As shown in Column 1, lines 44-68, Lowe et al. is merely concerned with making zeolite with a reduced amount of impurities. That portion of Lowe et al. is replicated below:

Zeolites and other similar zeolitic materials are usually prepared by crystallisation from aqueous reaction mixtures containing sources of silica and, where necessary, alumina. Many such reaction mixtures are prepared using strong bases, particularly alkali metal and quaternary ammonium hydroxides although weak bases are sometimes used in other cases. The nature of the quaternary ammonium and/or other organic base used as well as the mole ratios of the various reactants in the reaction mixtures have considerable influence on the zeolite or zeolitic material which is obtained. It is not unusual to obtain more than one zeolitic product from a reaction mixture and it may also [sic] happen that non-zeolitic crystalline materials and/or amorphous products are obtained. The preparation, therefore of a sufficiently pure zeolitic product in good yield is a problem constantly encountered by those in the zeolitic art. The Applicants believe that the method of this invention will help to reduce the difficulties inherent in the preparation of zeolites and zeolitic material. [Emphasis added]

Accordingly the present invention provides a method for the preparation of a crystalline zeolite or zeolitic material which comprises reacting an aqueous mixture comprising at least one source of silica and an ion exchange resin.

As shown above, Lowe et al. is only concerned with the preparation of zeolite without impurities. Lowe et al. is not concerned with making the macrostructures claimed in Claims 1 through 41. In fact, Lowe et al. in Column 3, lines 17-33, mentions that the Lowe et al. method is advantageous in forming the zeolitic material of U.S. Patent 4,073,865 and U.K. Patent Spec. No. 2,084,552 (copies enclosed). Both U.S. Patent 4,073,865 and U.K. Patent Spec. No. 2,084,552 form crystals of a silica polymorph, not macrostructures.

Further evidence showing that Lowe et al. is only concerned in the preparation of zeolite without impurities is set forth shown in Column 2, lines 9-14, which is replicated below:

The Applicants believe that when the aqueous reaction mixture contains an anion-exchange resin the changes on pH that commonly occur during zeolite synthesis are much reduced because the resin provides buffer capacity to cope with the changes in hydroxide ion concentration through its exchange equilibria.

In Column 2, lines 56-58, of Lowe et al. also shows that Lowe et al. does not make macrostructures. Lowe et al. recovers the resin used to make the zeolitic material. That portion of Lowe et al. is replicated below:

The ion-exchange resin is readily separated from the zeolitic product and easily regenerated with cheap aqueous alkali metal hydroxide

In contrast, the porous organic ion exchanger used to make Applicants' macrostructures is not recovered, but is destroyed. The inorganic oxide macrostructures of Applicants' invention occupy the internal pore space of the porous organic ion exchanger. Lowe et al. can recover resin, because the zeolitic material of Lowe et al. is not formed within the resin.

It is submitted that Lowe et al. is only concerned with making zeolitic crystals and does not disclose or suggest Applicants' claimed invention. Withdrawal of this rejection is respectfully requested.

Smith et al. does not cure the deficiencies of Lowe et al. Smith et al. is only concerned with hydrocarbon conversion using large crystal zeolite. Smith et al. discloses the use of a binder to bind together its zeolite particles. Binder material is not required as Applicants' macrostructures are made up of self-binding particles. Withdrawal of this rejection is respectfully requested.

REJECTION BASED ON OBVIOUS-TYPE DOUBLE PATENTING

Claims 1-44 stand rejected over the pending claims of U.S. Application Serial No. 09/574,432. The rejection is respectfully traversed.

In the interest of expediting the prosecution of the present application, enclosed herewith is a Terminal Disclaimer for U.S. Application Serial No. 09/574,432.

Withdrawal of this rejection is respectfully requested.

CONCLUSION

In conclusion, it is submitted that Lowe et al. either alone or in combination with Smith et al. does not disclose or suggest Applicants' claimed invention.

Respectfully submitted,

August 21, 2003
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